

Appendix A
Preliminary Release Rate Modeling Results

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To estimate the release rate of radionuclides from low-level waste disposal facilities, Brookhaven National Laboratory developed the Disposal Unit Source Term (DUST) model in support of the U.S. Nuclear Regulatory Commission. The Idaho National Engineering and Environmental Laboratory (INEEL) is currently using DUST to estimate radionuclide release rates from the SDA as input for the fate and transport simulator used in the Waste Area Group 7 baseline risk assessment.

A one-dimensional finite difference model is used to estimate the release. In DUST, the transport equation is solved with the processes of advection, diffusion, retardation, and radioactive decay. The variety of modeled processes allows the comparison of in situ grouting (ISG) release (diffusion controlled) and baseline release (advection controlled). For illustrative purposes, release rates were calculated for two different infiltration rates to simulate baseline conditions and a low permeability cap, and for ISG.

Release from many solid waste forms can be described as a diffusion-controlled process (Sullivan 1993). The DUST model analytically solves the diffusion equation, corrected for decay.

$$\frac{\partial C}{\partial t} = \nabla \cdot D \nabla C - \lambda C \quad (1)$$

where

C = solution concentration

D = the diffusion coefficient

λ = radioactive decay constant.

Mass flux at the surface, for one-dimensional diffusion controlled release, is:

$$J_s = -D \frac{\partial C(x_s)}{\partial x} \quad (2)$$

where

x_s = a surface of the waste form.

Finally, the mass flux is integrated over the surface area to yield the release rate.

$$Q(t) = \int dS \cdot J_s \quad (3)$$

where

$Q(t)$ = the mass release per unit time

J_s = the mass flux at the surface.

To simplify the situation, the concentration in the contacting solution is assumed to be zero. This assumption leads to the highest predicted release rates (conservative) and permits an analytical solution. The initial condition assumes a uniform concentration throughout the waste form. Symmetry is assumed about the midplane of the waste form and zero concentration at the outer edge. Because cracking frequency and aperture data are not available, the source term is partitioned into 1-m³ blocks. Conceptually, this represents the size of the grout monolith between hypothetical, worst-case fractures. The contaminants diffuse slowly through each 1-m³ block but are immediately washed away as they reach the edge of the waste form.

Parameters used in the model runs are shown in Table A-1.

Table A-1. Parameters used in the model run.

Parameter	Baseline	Low Permeability Cap	Grouted Waste Form
Waste form geometry	Rectangular; 13 areas 14.2 ft thick	Rectangular; 13 areas 14.2 ft thick	Rectangular; 1 × 1 × 1 m
Infiltration rate	Baseline rate from lysimeter measurements	1 cm/year	1 cm/year
Release mechanism	Surface wash off	Surface wash off	Diffusion
Partition coefficient ^a	U-6	U-6	
	Np-8	Np-8	
	Am-450	Am-450	
Diffusion coefficient ^b	—	—	U-10 ⁻¹³

a. Dicke 1997

b. Weidner et al. 2000

Solubility limits are not considered in this exercise. The DUST model sets a boundary condition of zero concentration at the waste form solute interface. Changes in the distribution coefficient resulting from changes in pH and Eh, as well as competition with other ions for sorption sites, are not considered. Because the actual SDA water is saturated with minerals (e.g., calcite) (Weidner et al. 2000), and because the redox and pH of candidate grouts are designed to minimize solubility of certain contaminants (Hull and Pace 2000), actual ISG release rates are expected to be significantly lower than those calculated by the DUST model.

The DUST model provides a quick method of computing release rates and is useful for screening, sensitivity analysis, and (with confidence in input parameters) can provide upper bound release rates. In using the DUST model or any other analytical solution to estimate release rates, the largest uncertainties are attributable to uncertainties in the input parameters. In this exercise, the problem has been simplified by making a number of assumptions. However, to accurately and defensibly estimate release rate, contaminant-specific and grout-specific diffusion data are required.

Figures A-1 and A-2 plot the results of the DUST model runs for two radioisotopes, U-234 and U-238. Additional runs for other contaminants of interest could not be completed at this time because of unavailable diffusion coefficient data or unavailable contaminant inventories.

Release rates in milligram per year are plotted over years, and the model is run over 10,000 years. The rate of diffusion is relatively constant beyond the 4,000-year point. As expected, the shape of the curves for both isotopes is similar. The difference in release rates between the two is attributed to the different starting inventories. The apparent spikes in the earliest time steps of the ISG curve may be attributable to an artifact of the diffusion equation where the release rate approaches infinity as time approaches zero (Sullivan 1993). For the base case and cap data, the initial spike near time zero may be attributable to the fact that the model starts release rates at zero but then overcorrects in an effort to achieve mass balance. The release rates for all three cases quickly stabilize and slowly decay over time. This exercise demonstrates clearly that the release of uranium (and other contaminants with similar diffusion coefficients in cementitious waste forms) is nearly two orders of magnitude less than the base case and the case of a low permeability cap.

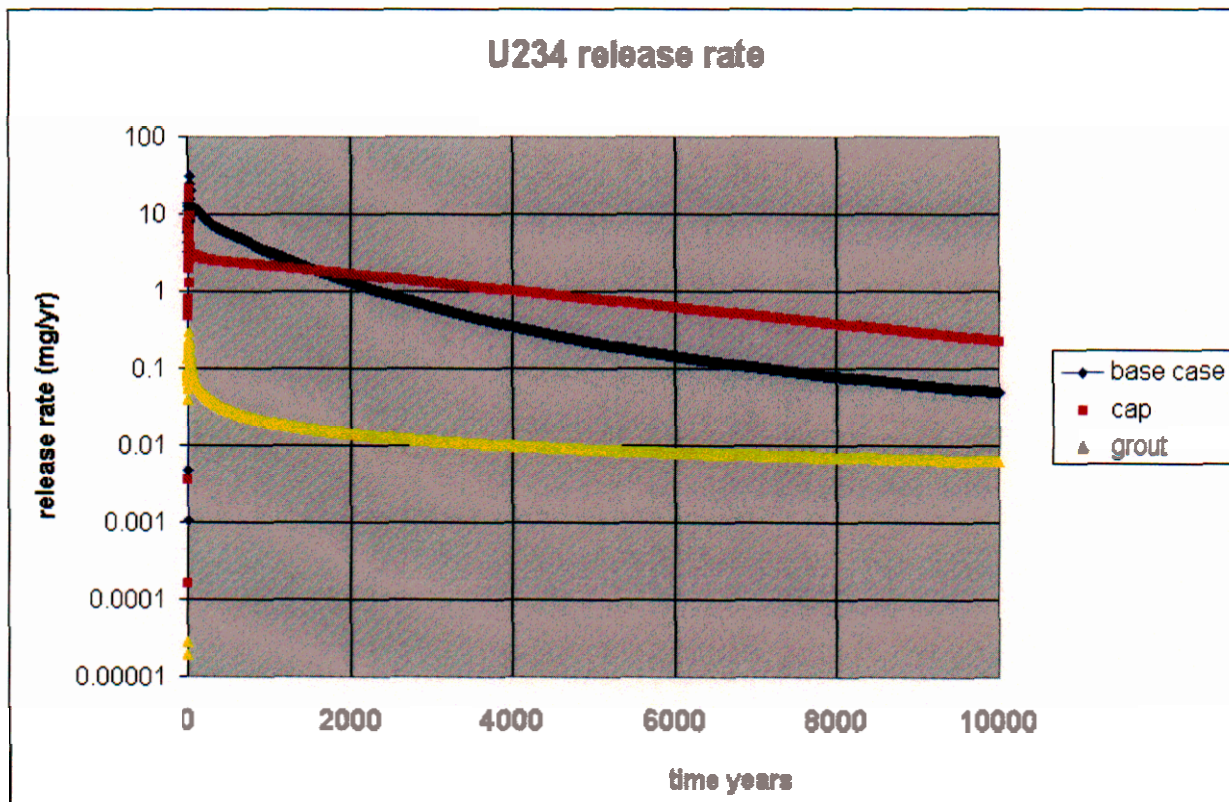


Figure A-1. Uranium-234 release rates as estimated using the Disposal Unit Source Term model.

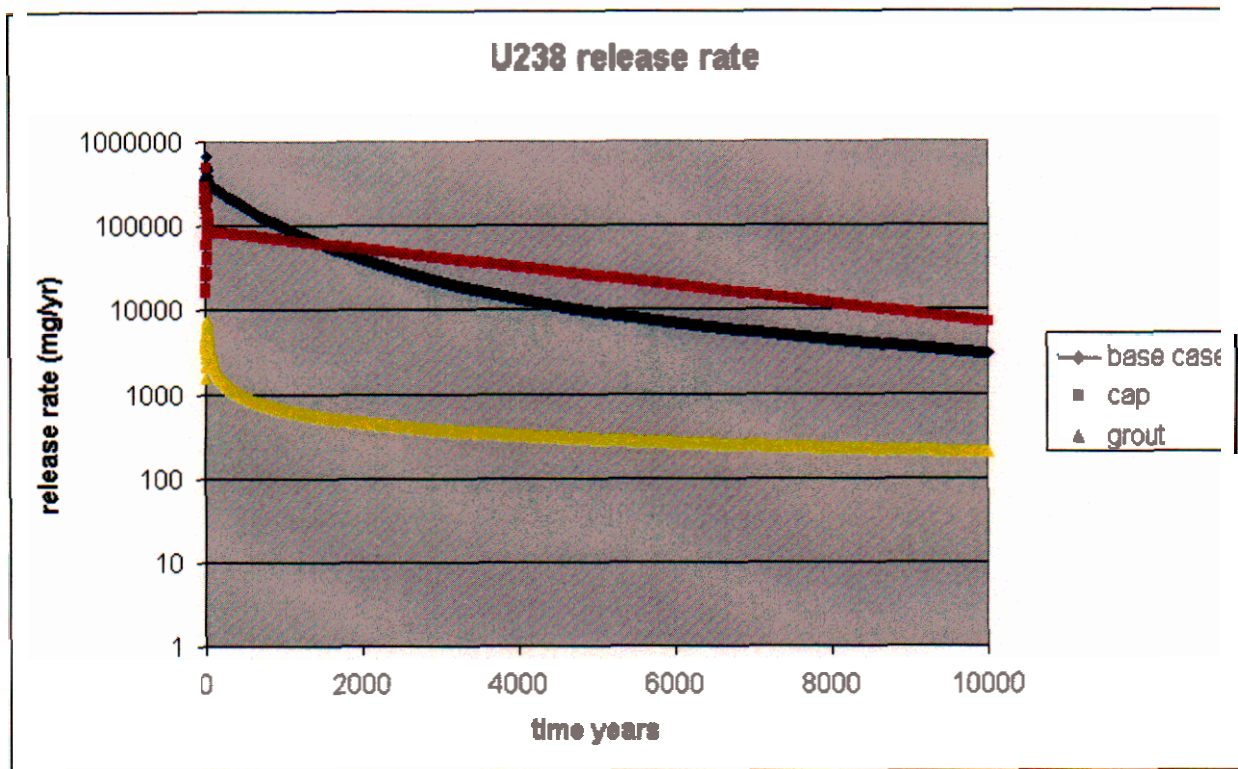


Figure A-2. Uranium-238 release rates as estimated using the Disposal Unit Source Term model.

References

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- Hull, Laurence, and Mary N. Pace, 2000, *Solubility Calculations for Contaminants of Potential Concern at OU 7-13/14*, INEEL/EXT-2000-00465, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.
- Weidner, Jerry, Paul Blacker, Mary Pace, and Laurence Hull, 2000, *Biodegradation of Grout, Contaminant Diffusion, Solubility, and Technical Review of the In Situ Grout Treatability Study*, Chapter 3, "Contaminant Diffusion Coefficient Data," INEEL/EXT-2000-00511, Rev. 0, prepared by CH2MHILL for the Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.
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